Dimensional crossover and anomalous magnetoresistivity in single crystals Na_xCoO_2

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The in-plane (ρ_{ab}) and c-axis (ρ_c) resistivities, and the magnetoresistivity of single crystals Na_xCoO_2 with x = 0.7, 0.5 and 0.3 were studied systematically. $\rho_{ab}(T)$ shows similar temperature dependence between $Na_{0.3}CoO_2$ and $Na_{0.7}CoO_2$, while $\rho_c(T)$ is quite different. A dimensional crossover from two to three occurs with decreasing Na concentration from 0.7 to 0.3. The angular dependence of in-plane magnetoresistivity for 0.5 sample shows a *"d-wave-like"* symmetry at 2K, while the *"p-wave-like"* symmetry at 20 K. These results give an evidence for existence of a *spin ordering orientation* below 20 K turned by external field, like the stripes in cuprates.

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Recently the study of layered sodium cobaltate oxide Na_xCoO_2 has became one of the hot topics in research of condensed matter physics. Na doping leads to the change of spins from spin 1/2 for Co^{4+} to spinless for Co^{3+}. The discovery of superconductivity with $T_c \sim$ 5 K in $Na_{0.35}CoO_2 \cdot 1.3H_2O$[1] makes one consider that Na_xCoO_2 may be a system like cuprates where superconductivity occurs in a doped Mott insulator. Furthermore, it is also expected that the triangle lattice based cobaltates may exhibit some novel electronic and magnetic phases, for example, the Anderson's RVB states[2] and the strong topological frustration phases[3, 4, 5]. The in-plane magnetic correlation of Na_xCoO_2 is complex and still not clear. The ^{59}Co nuclear quadrupolar resonance measurement suggests a two-dimensional antiferromagnetic correlation in $Na_{0.35}CoO_2 \cdot yH_2O$[6]. However, the band structure calculations predict a ferromagnetic spin fluctuations within the CoO_2 plane for Na_xCoO_2[7, 8]. The existence of in-plane ferromagnetic correlations is confirmed by inelastic neutron scattering experiments for x=0.75[9] and x=0.82[10] crystals.

Besides the magnetic properties, Na_xCoO_2 system also shows many anomalous transport properties. Extremely large and magnetic field dependent thermoelecreic power (TEP) is observed. The enhancement of TEP is believed to be due to spin entropy in $Na_{0.68}CoO_2$ [11]. The Hall coefficient is found to be linear temperature-dependent in a wide temperature range and shows no saturation up to 500 K[12], while in a conventional metal Hall coefficient is temperature independent. The unusual linear-T resistivity is found in low temperatures[11, 13, 14] showing non-Fermi liquid behavior. While observation of a T^2 dependence of resistivity at ultra low temperature and the satisfaction of the Wiedemann-Franz law in $Na_{0.7}CoO_2$ show the validity of ordinary Fermi-liquid state in ultra-low temperatures with enormous e-e sacttering[15]. In addition, a so-called "incoherent-coherent" transition with decreasing temperature is observed in $\rho_c(T)$. ARPES experiments reveal that well-defined quasiparticle peaks develop only at low temperature in $Na_{0.75}CoO_2$ when a dimensional crossover occurs[14, 16]. The electronic and magnetic properties in Na_xCoO_2 is very sensitive to the Na content[17]. Therefore, it would be helpful to study the evolution of transport properties with doping level in this system.

The anisotropic transport measurement is a useful way to the understanding of electronic and magnetic properties such as the dimensional transition. Magnetoresistance measurement is also a useful tool to get further insight into the anomalous charge transport since it is more sensitive to the change in the charge carrier scattering rate $1/\tau$, effective mass m^* and the geometry of the Fermi surface. In this letter, the anisotropic transport properties of Na_xCoO_2 single-crystals are studied systematically. A dimensional crossover from two to three occurs with decreasing the Na content. The angular dependence of the in-plane resistivity for $Na_{0.5}CoO_2$ shows a two-fold symmetry at 20 K, while four-fold symmetry at 2 K. Such anomalous angular dependence of MR is related to a special *"spin ordering orientation"*, such as the stripes, which can be turned by the applied field.

High quality single crystals $Na_{0.7}CoO_2$ were grown using the flux method. The typical dimensional is about $2 \times 1.5 \times 0.01mm^3$ with the shortest dimension along the c axis. The x=0.5 and 0.3 samples were achieved by Na deintercalation of $Na_{0.7}CoO_2$ in solutions of iodine- and bromine-dissolved acetonitrile. The actual Na concentration is determined by inductively coupled plasma analysis (ICP) experiments. No impure phase is detected within experimental error of 2% in x-ray diffraction measurements. The c-axis lattice parameter is about 10.960, 11.115 and 11.215 $\mathring{A}$ for x=0.7, 0.5 and 0.3, respectively. This is consistent with the data reported by Foo et al[13]. To obtain the coincident results, one single crys-

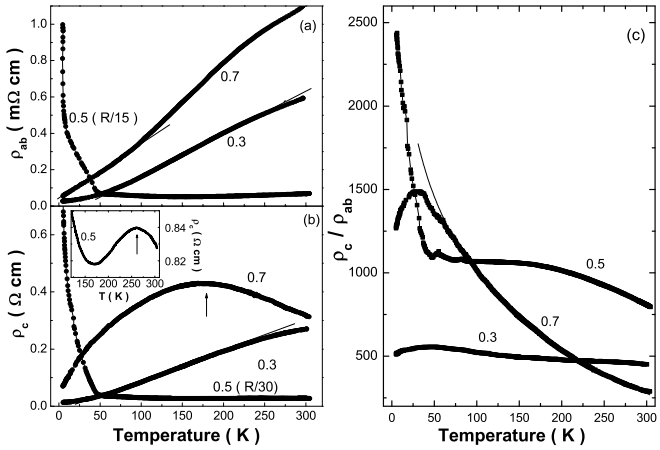

FIG. 1: Temperature dependence of (a) in-plane resistivity (ρ_{ab}); (b) out-of-plane resistivity (ρ_c), and the thin line guides for eyes; (c) temperature dependence of anisotropy (ρ_{ab}/ρ_c) for Na_xCoO_2 with x=0.7, 0.5, and 0.3, the line is the fitted result by $ln(T_0/T)$ for x=0.7 sample.

tal was cut into two pieces. One is for in-plane resistivity measurement and another for out-of-plane measurement. The resistivity and magnetoresistance were performed in Quantum Design PPMS systems.

$\rho_{ab}(T)$ and $\rho_c(T)$ for Na_xCoO_2 with x=0.7, 0.5, and 0.3 are presented in Fig. 1(a) and (b). As reported by other groups[11, 13, 14], a T-linear behavior of $\rho_{ab}(T)$ is observed in $Na_{0.7}CoO_2$ in low temperatures below 100 K, indicating non-Feimi-liquid-like behavior[14]. In the temperature range between 100 and 180 K, $\rho_{ab}(T)$ can be well fitted by a $T^{3/2}$ law. Above 180 K, $\rho_{ab}(T)$ follows another power-law (T^{α}, $\alpha < 1$) behavior. It is worth noting that the $T^{3/2}$-dependent behavior has been reported by Rivadulla et al.[19] in several metallic systems including Na_xCoO_2. They argued that this phenomenon presents in metals which approach the Mott-Hubbard transition from the itinerant-electron side, and concluded that locally cooperative bond-length fluctuations at isolated clusters can account for this behavior. $\rho_{ab}(T)$ for x=0.5 sample shows a weak metallic behavior above 160 K, then a weak insulating behavior as T falls toward 50 K. Below 50 K, $\rho_{ab}(T)$ grows sharply, indicating the opening of charge ordering gap at about 50K. $\rho_{ab}(T)$ increases more sharply below 20 K. The result is consistent with that observed by Foo et al[13].

We have carefully analyzed the data of 0.3 sample. It is found that the T^2 behavior is observed in $\rho_{ab}(T)$ below about 30 K as previous report[13]. A $T^{3/2}$ temperature dependence is observed from 30 to about 100 K, then a T-linear behavior between about 100 and 220 K. Above 220 K, $\rho_{ab}(T)$ can be described by T^{α}' with α'<1 as the case of $Na_{0.7}CoO_2$. In comparison with x=0.7 sample, $\rho_{ab}(T)$ for 0.3 sample shows also four different temperature dependence regimes. But the T^2 dependence of resistivity occurs below 30 K, which is much higher than that (1 K) observed in the 0.7 sample[15]. In addition, the T-linear behavior begins to show up at 220 K, while at 100 K in the 0.7 sample. Although $\rho_{ab}(T)$ experiences similar temperature dependence with increasing temperture ($T^2 \rightarrow T^{3/2} \rightarrow T \rightarrow T^{\alpha}$'($\alpha$'<1)) to the case of $Na_{0.7}CoO_2$ ($T^2 \rightarrow T \rightarrow T^{3/2} \rightarrow T^{\alpha}(\alpha < 1)$), $\rho_c(T)$ shows quite different behavior.

The temperature dependences of ρ_c for the three samples are shown in Fig. 1(b). $\rho_c(T)$ shows a so-called "incoherence-coherence" transition peak at T_M around 180 K for $Na_{0.7}CoO_2$ sample and 260 K for $x = 0.5$, respectively, similar to the report by Valla et al[16, 20] in $(Bi_{0.5}Pb_{0.5})_2Ba_3Co_2O_y$ (T_M ~200 K) and $NaCo_2O_4$(T_M ~180 K). They considered it as a crossover in the number of effective dimensions from two to three. However, such maximum in $\rho_c(T)$ is not observed in the 0.3 sample over the whole temperature range. As observed in $\rho_{ab}(T)$ for x=0.3 sample, $\rho_c(T)$ also shows a T^2 dependent behavior in the temperatures below 30 K, and T-linear behavior from 100 to 220 K. This indicates that the x=0.3 sample has the same temperature dependence for both $\rho_{ab}(T)$ and $\rho_c(T)$, suggesting the same scattering mechanism. It may explain why no "incoherence-coherence" transition peak is observed. While for x=0.7 sample, the temperature dependence of $\rho_{ab}(T)$ is completely different from that of $\rho_c(T)$. It suggests that there exists different scattering mechanism between in-plane and out-of plane charge transports. For $Na_{0.5}CoO_2$, the $\rho_c(T)$ shows an "incoherence-coherence" transition peak at 260 K, and a metallic behavior is observed from 260 K to 165 K. Below 165 K, it shows a similar behavior as observed in ρ_{ab}, but the insulating behavior is much sharper than that in ab-plane. It should be pointed out that the gross feature in ρ_{ab} and ρ_c is similar to each other. In one word, $\rho_{ab}(T)$ and $\rho_c(T)$ show the same scattering mechanism for $Na_{0.3}CoO_2$, while different scattering mechanism for $Na_{0.7}CoO_2$. The temperature corresponding to "incoherent-coherent" transition increases with decreasing Na content. These results suggest a dimensional crossover from two to three with decreasing Na concentration from 0.7 to 0.3, being similar to the observation in $La_{2-x}Sr_xCuO_4$ with increasing x[18]. In both of the cases, the carrier increase leads to a dimensional crossover from two to three companied with the decrease in spin correlation.

$\rho_c(T)/\rho_{ab}(T)$ is plotted in Fig. 1(c). $\rho_c(T)/\rho_{ab}(T)$ shows weak temperature dependence for the x=0.3 sample. However, $\rho_c(T)/\rho_{ab}(T)$ increases with lowering temperature and has strong temperature dependence above 50 K for the x=0.7 sample, and it can be well fitted by $ln(T_o/T)$ above 50 K. $\rho_c(T)/\rho_{ab}(T)$ shows also a weak temperature dependence above 165 K for the x=0.5 sample, and a saturation between 165 and 50 K, while below 50 K the anisotropy increases quickly, implying that the insulating behavior of ρ_c is stronger than ρ_{ab} due to charge ordering. These results further indicate that

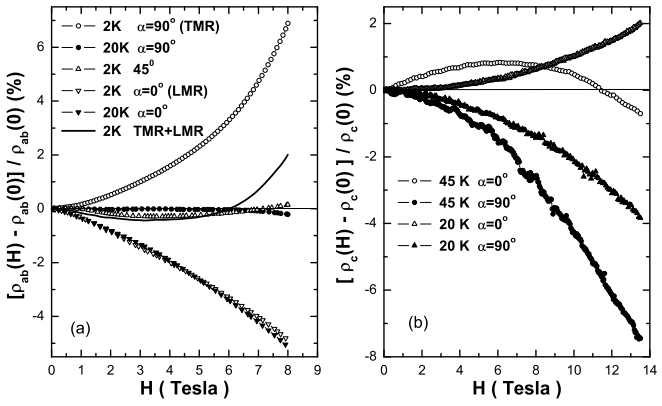

FIG. 2: In-plane and out-of-plane magnetoresistivities with different angles between H and I for $Na_{0.5}CoO_2$ at 2, 20, and 45 K, respectively. The line in (a) is the result of TMR plus LMR at 2 K.

a dimensional crossover from two to three occurs with decreasing Na content. It should be addressed that although ρ_c for the 0.5 and 0.7 samples show a "incoherent-coherent" transition peak at 260 and 180 K, respectively, the ρ_c/ρ_{ab} does not show any anomaly at the peak temperature. The anisotropy shows a maximum at about 40 K for x=0.7 sample, which is much lower than the so-called coherence-incoherence transition (180 K). For the 0.5 sample, the anisotropy increases slightly with decreasing temperature, and shows a saturation at around 165 K which coincides with the crossover from metallic to nonmetallic behaviors in ρ_{ab} (165 K). It should be pointed out that the low temperature anisotropy maximum for the x=0.7 and x=0.3 samples corresponds to the temperatures at which the in-plane thermal conductivity κ and Hall coefficient R_H begin to slightly drop for both $x = 0.71$ and 0.31[13]. It suggests that the anisotropy maximum is correlated with the change in the charge transport character.

Magnetoresistivity ($MR = [\rho(H) - \rho(0)]/\rho(0)$) was measured by sweeping the field at fixed temperature. Variation of the in-plane and out-of-plane MR with H at fixed temperatures for x=0.5 sample are shown in Fig. 2. Figure 2(a) shows the in-plane MR at 2 and 20 K under the fields with different angles relative to ab plane. A large, negative MR is observed at 20 K with H in ab plane and along I, while the MR is very small with $H \perp ab$ plane. At 2 K, a large, positive MR is observed with $H \perp ab$ plane, while a large, negative MR is observed when H is applied within ab plane and parallel to I. However, the MR shows a complicated behavior with an angle of 45 degree between H and I, the MR is negative, and monotonically increases with H up to 3.5 T, then decreases to zero at about 7 T and changes the sign with further increasing H. It implied that the MR is contributed by a positive and a negative component. In order to understand it, we calculate the result of MR with $H \perp ab$ adding MR with $H \parallel ab$, shown as the solid line in Fig. 2(a). The calculated result is almost the same as the experimental one except for the high H regime. It further suggests that the MR with an angle of 45 degree between H and I is a competing result between the MR's with $H \perp ab$ plane and $H \parallel ab$ plane, respectively. Usually, the transverse MR contains the orbital contributions, involving the contribution from the "bending" of electron trajectory by the Lorentz force, which is always positive. But here the positive MR is not only from the "bending" of electron trajectory, the main contribution should be from spin related effect because the transverse MR at 20 K is almost zero. If the transverse MR would originate from the conventional orbital motion of carriers, the MR should follow the Kohler's rule, i.e. $\Delta\rho_H/\rho_0$ vs $(H/\rho_0)^2$ forms a single universal relation, independent of temperature[24]. However, the MR at 2 K and 20 K violates the Kohler's rule obviously. In addition, the longitudinal MR at 20 K is nearly the same as that at 2 K. Because low temperature MR is usually larger than that at high temperature except for a phase transition, it further suggests that the MR is dependent on spin ordering in low temperatures, spin reorientation at 20 K[23].

The isothermal out-of-plane magnetoresistivity is shown in Fig. 2(b). At 45 K, the longitudinal MR shows a complicated behavior, and experiences a maximum at about 6 T, while the transverse MR monotonically increases with H. At 20K, the longitudinal MR is positive and increases with increasing H, a negative transverse MR is observed. This behavior is similar to that of in-plane at 2 K shown in fig. 2(a). It is found that both the in-plane and out-of-plane MR below 20 K is positive with $H \perp ab$ plane, while negative with H applied in ab plane. Another interesting feature is that the transverse MR at 45 K is much larger than that at 20 K. It is considered that the larger negative MR could arise from the suppression of charge ordering at about 50 K by H[25]. These results have indicated that the MR is strongly dependent on spin ordering. μSR measurements by Uemura et al. gave an evidence that an antiferromagnetic order sets in at the onset of a metal-insulator transition in the x=0.5 system. They gave a possible picture that one of two interpenetrating Co spin networks acquires a long-range order below 53 K, followed by the other network established long-range order below 20 K[22]. At this moment, the spin structure in the x=0.5 system is not clear, a conclusive picture requires neutron scattering studies. Therefore, it is difficult to clearly explain the observed MR behavior. The MR results seem to give an evidence that the spin correlation is ferromagnetic in ab planes, while antiferromagnetic between ab planes.

In order to understand the strong field direction dependence of the MR observed in Fig. 2, the detailed in-plane MR measurements are performed upon rotating H of 8 T, and H, I and c-axis always are kept in the same plane. Evolution of the MR with the angle between H and I is shown in Fig. 3(a). In-plane MR at 20 K is negative over

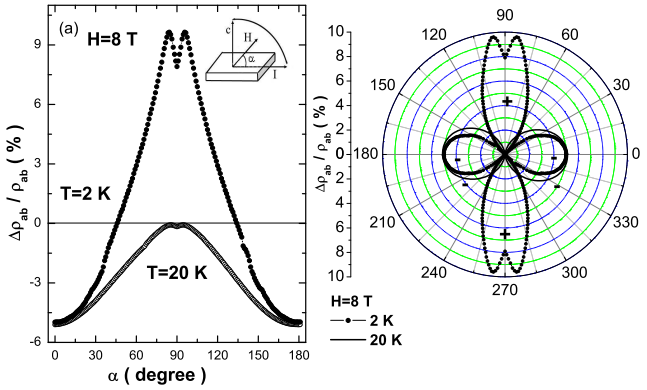

FIG. 3: The angular dependence of in-plane magnetoresistivity for $Na_{0.5}CoO_2$ at H=8 T. The angle between H and I is changed upon rotating H. H, I and c-axis are kept in the same plane when the field is rotated.

all angle range. At $\alpha = 0$ (α is the angle between H and I), the in-plane MR is maximum, while a MR is almost zero at $\alpha = 90^0$. A quite different behavior shows up at 2 K, a striking anisotropy with a *"d-wave-like"* symmetry is observed in Fig. 3(b). $\Delta\rho_{ab}/\rho_{ab}$ changes from negative at $\alpha = 0^o$ to positive at $\alpha = 90^o$, passing through zero at $\alpha = 45^o$. It should be noted that at 2 K the positive term extending the "+" arm at $\alpha = 0^o$ is larger relative to the "-" arm at $\alpha = 90^0$. Angular dependent MR at 20 K is also plotted in pole figure shown in Fig. 3(b), the MR shows a two-fold symmetry. It should be pointed out that the MR diagram in Fig. 3 is fairly symmetric at 20 and 2 K. The common behavior at the two temperatures is that the negative arm appears at $\alpha = 0^o$ and 180^o, while the difference is the absence of the positive arm at 20 K. It suggests that the spin structure is changed with temperature decreasing from 20 to 2 K, consistent with that observed by μSR [22], leading to the resistivity change at 20 K shown in Fig.1(a) and (b). In x=0.5 system, the long-range antiferromagnetic order occurred in two different Co spin networks at 53 K and 20 K, respectively[22], or the spin reorientation took place at 20 K[23]. So far, we cannot give an exact explanation about the anomalous "d-wave" shaped angular dependent MR due to the unknown spin structure. A similar "d-wave-like" symmetric MR was observed below the Neel temperature in antiferromagnetic $YBa_2Cu_3O_{6+x}$[26] and $Nd_{2-x}Ce_xCuO_4$[27]. In $YBa_2Cu_3O_{6+x}$, the magnetic field would give rise to a topological ordering of the stripes, aligning them along the field direction and changing the array of the current paths[26]. While in Nd_2CuO_4 the spins prefer to be perpendicular to the field when the external field is applied, so that the spin structure changes from noncollinear to collinear[28]. In both of the cases, the *"d-wave"*-like anisotropy of in-plane MR is understood as a consequence of the rotation of the stripe direction with respect to the current direction[26] and of the collinear spin direction relative to the current[27]. We have no evidence for existence of a special spin structure in the x=0.5 system, like the "spin stripes" ordering in $YBa_2Cu_3O_{6+x}$ or "collinear spin structure" in Nd_2CuO_4, but the anomalous angular dependence of MR implies existence of a *"spin ordering orientation"* turned by the field. Therefore, neutron scattering measurement is acquired to resolve this question.

In summary, transport properties and magnetoresistance were studied in Na_xCoO_2 ($x = 0.3$, 0.5, and 0.7). It is found that $\rho_{ab}(T)$ shows similar temperature-dependent behavior in $Na_{0.7}CoO_2$ and $Na_{0.3}CoO_2$, while $\rho_c(T)$ shows different behavior. A dimensional crossover from two to three also takes place with decreasing Na concentration. The angular dependent in-plane magnetoresistivity for the x=0.5 system is a *"d-wave-like"* symmetry at 2 K, while a *"p-wave-like"* symmetry at 20 K. Such anomalous angular dependence of MR is related to a special *"spin ordering orientation"* turned by the applied field. Neutron scattering measurement under the external field is needed to resolve this issue

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